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Amorphous Phase Separation, Salt Precipitation, and High Pressure

Effects in PPO containing NaI

by

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AMORPHOUS PHASE SEPARATION, SALT PRECIPITATION, AND HIGH PRESSURE
EFFECTS IN PPO CONTAINING NaI

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ABSTRACT

Audio frequency dielectric constant and electrical conductivity, ^{23}Na NMR, and differential scanning calorimetry studies have been carried out on poly(propylene oxide) (PAREL elastomer) containing various concentrations of NaI. Both the NMR and the electrical conductivity measurements were performed at high pressures. The low temperature DSC results show the existence of two amorphous phases, one very dilute and one with a concentration of approximately 8 monomer units per sodium. All techniques applied indicate that the more concentrated phase is the ion conducting phase. At high temperatures, however, the conductivity decreases as temperature increases. The decrease coincides with an endotherm in the DSC studies which is associated with salt precipitation. Further, the DSC studies show that the salt precipitation temperature decreases as the concentration increases. An explanation for the salt precipitation phenomenon is suggested in that it is found that the dielectric constant of the host polymer decreases as temperature increases. The NMR measurements reveal the presence of both bound and mobile sodium species, where the bound phase probably corresponds to small salt clusters. All high pressure results suggest an increase in the glass transition temperature with applied pressure.

INTRODUCTION

Poly(propylene oxide) (PPO) has been studied extensively in the past few years. The primary reason is that it is easy to prepare amorphous PPO salt complexes with relatively high electrical conductivity and thus the results are free of the complications due to crystallinity such as exist for poly(ethylene oxide). One of the important issues in polymer electrolyte science involves an understanding of ion-ion interactions which are expected to be substantial in view of the high salt concentrations in these materials. One feature of these materials which is yielding a great deal of information concerning ion-ion interactions is that at high temperatures sodium and potassium salts tend to precipitate out of the polymer in a dramatic fashion (1-4). Previous work on PPO_xNaI (3)

focused on the particular manner in which the salt precipitation (SP) resulted in anomalies in differential scanning calorimetry (DSC), ^{23}Na nuclear magnetic resonance (NMR), and electrical conductivity measurements at higher temperatures. In this paper, studies of the dielectric constant of the host polymer are reported. Those results give insight into the reason that SP occurs. In addition, the effects of salt concentration are studied by investigating values of $n = 8, 12,$ and 16 , where n is the oxygen/sodium ratio of the complex. Next, for different salts in PPO, a correlation between the SP temperature and the melting point of the salt is pointed out. Both effects on the SP temperature, salt concentration and species, are discussed in terms of ion-ion and ion-polymer interactions. Next, the low temperature DSC studies reveal a previously unreported aspect of the morphology of these materials. Finally, high pressure conductivity and ^{23}Na NMR data are also reported.

EXPERIMENTAL

The samples were prepared using solution casting with methanol as the solvent. All preparations were done in a dry box with final drying done in a vacuum oven with an initial anneal at 110°C for 24 hours and a second anneal at about 60°C for at least 24 hours. The samples studied in the present work were stored over desiccant for several months before any measurements were performed.

The techniques employed in the DSC, audio frequency dielectric relaxation, and electrical conductivity studies are described elsewhere (2,3,5-10). The ^{23}Na NMR measurements were performed at 81 MHz utilizing standard pulse techniques. Good signal to noise ratios required averaging of typically 100-150 repetitions. For the high pressure measurements, the sample was enclosed in a rubber finger cot in order to isolate it from the pressure transmitting fluid (Fluorinert FC-77), and then loaded into the rf coil inside the pressure vessel. The entire bomb was heated to 45°C by a warm air stream and allowed to reach thermal equilibrium for about 1 hour prior to the measurement.

RESULTS

Amorphous Phase Separation

The results of the low temperature DSC runs are given in Fig. 1a. Two glass transitions are observed in all samples containing NaI, one in the vicinity of that for uncomplexed PPO (approximately 210°K) and one in the vicinity of 270°K . The effect of increasing the concentration of the salt is merely to increase the strength of the higher temperature glass transition at the expense of the lower temperature glass transition. Consequently, there is a tendency for the ions to form an amorphous phase with a well-defined stoichiometry. As very little low temperature glass transition is observed for

PPO₈NaI, it is concluded that the concentration of the well-defined phase is close to 8:1 (though it may contain slightly more salt). Figure 1b displays the high temperature DSC data for three samples. The relatively sharp endotherms are attributable to SP as will be discussed later. The preference for a particular stoichiometry is in contradiction to what is usually observed in these materials in that the salt can, in most cases, assume any concentration with the glass transition temperature increasing as concentration increases.

However, this result is also consistent with the present NMR results. A general feature of the NMR in sodium-conducting polymers is the presence of two distinct spectral responses with substantially different spin-lattice relaxation times (T_1) (2,3,8). The longer T_1 component of the signal does not show a pronounced temperature dependence of either T_1 or its linewidth. The shorter T_1 component, on the other hand, exhibits substantial motional narrowing above T_g . Figure 2 displays the two separate components of the free induction decay (FID) in PPO₈NaI at 56C, obtained by selective saturation and subtraction. The top FID corresponds to a T_1 of about 2 ms and is partially narrowed, as evidenced by the relatively long decay. The bottom FID has a T_1 of about 10s. These two components have previously been attributed to bound (long T_1) and mobile (short T_1) phases of sodium in the complex (2,3,8). The temperature insensitivity of the bound Na signal, even above the SP temperature (to be discussed later) suggests that the bound Na may, in fact, exist in the form of small clusters (>10 ions) of salt that are spectroscopically similar to pure NaI (both in linewidth and T_1) but too small to be seen by x-ray diffraction below the SP temperature. One possibility for the placement of these ions is at the boundary between the phases. This is discussed further below. Contact ion pairs or triplets would be expected to yield a broader line than observed, in light of substantial nuclear quadrupole couplings deduced from gas phase measurements (8). The NMR results do not rule out the presence of ion pairs or triplets if, for example, they exist in equilibrium with single ions and larger clusters. If this is the case, however, the NMR indicates that the relevant exchange rates are rapid enough to average out the quadrupole interaction (>1 MHz) characteristic of an ion pair.

The mobile ²³Na reciprocal FID time constants, $1/(T_2^*)$, (which are proportional to the linewidths) for all three PPO_nNaI complexes are plotted as a function of temperature in Fig. 3. Above room temperature the samples are in the motional narrowing regime, whereas above about 60C the resonances begin to broaden as a result of extremely fast T_1 's (T_1 about 300 microseconds) in this temperature region. The important feature of the data in Fig. 3 is that all samples exhibit approximately the same linewidth at a given temperature which indicates that the nature of the mobile phase of the sodium ions in the complex is independent of salt concentration, as suggested by the DSC results. The small and apparently systematic difference between the n=8 and n=12 and 16 linewidths probably results

from incomplete saturation of the bound Na signal in the latter two compositions. As discussed below, these materials have a substantially higher percentage of bound Na than $n=8$ and any residual signal from the bound Na results in an apparently shortened FID.

The temperature dependence of the ratio of mobile to bound Na (from integrated FID intensities) is plotted in Fig. 4. The lower temperature regions indicate that the $n=8$ composition has the highest percentage of mobile Na and consequently this represents the most homogeneous material. It is curious that the lower concentrations contain the largest percentage of bound sodium ions. In fact, this effect can be seen visually as the lower concentration materials are usually cloudier than the $n=8$ material. As mentioned above, this effect is explained if the ions or uncomplexed salt collect at the phase boundaries, the large ratio of mobile/bound sodiums in $n=8$ material being attributable to the lack of phase boundaries i.e. a more homogeneous material. As the temperature is increased the more dilute complexes exhibit a relatively rapid increase in mobile ion concentration attributable to a greater degree of cation solvation. This is not inconsistent with the DSC results as those data probably only reflect the morphology for room temperature and below. Finally, it is apparent from Fig. 5 that the $n=8$ sample exhibits the most rapid increase in electrical conductivity with increasing temperature while the number of mobile ions is relatively unaffected. Consequently, the NMR and the electrical conductivity data imply that the concentrated phase is a conducting phase. This is somewhat different from the results for poly(ethylene oxide) where it is generally assumed that the conducting phase is a dilute amorphous phase (where n is on the order of 16-20).

Salt Precipitation

As is apparent from Fig. 5, above about 80°C the electrical conductivity begins to decrease. This result is correlated with the high temperature DSC endotherms displayed in Fig. 1b and the sharp drop in mobile to bound Na ratio at high temperature shown in Fig. 4. Figure 1b shows that the $n=12$ and $n=16$ samples exhibit SP at higher temperatures than for $n=8$. This trend is also discernable in the electrical conductivity data of Fig. 5. These results can be explained if the salt precipitates out of the polymer in the vicinity of 80°C. Both the NMR and conductivity measurements imply lower SP temperatures than the DSC results, however, the samples were equilibrated for at least 30 minutes for NMR and conductivity, and scanned at 10K/min. for DSC. SP has been reported previously in PPO using a variety of techniques (1-4).

Before providing an explanation for this effect, a few comments concerning the electrical conductivity results are appropriate. The behavior is similar to that for PPO KSCN and is slightly different from the results for PPO NaI shown previously (3). In the case of PPO NaI, the conductivity showed a tendency to be overestimated at high

temperatures by the VTF equation fitted to the low temperature data, but the conductivity did not actually decrease. The explanation for the differences is that the former data were taken more rapidly, before all of the salt was allowed to come out of the polymer.

It remains to explain why salt precipitation occurs. In order to understand this result, the variation of the dielectric constant with temperature was investigated. The results for uncomplexed PPO are shown in Fig. 6. It is apparent that above the glass transition temperature the dielectric constant decreases with increasing temperature. The reason, of course, is that the dipoles which become "free to reorient" above the glass transition, represent a Curie-Weiss ($1/T$) contribution to the dielectric constant. Consequently, it is concluded that the salt precipitates out of the polymer when the dielectric constant becomes too low to mediate solvation. Clearly, this also depends upon the ion-ion interaction. In fact, data already exist to demonstrate the significance of ion-ion interactions. Specifically, it was shown above that the SP temperature increases with decreasing concentration. This result is expected because decreased concentration should decrease ion-ion interactions and hence increase the SP temperature. Further, it has been shown that for sodium salts, the SP temperature is about 80C, 140C, and 165C for iodide (3), perchlorate (2), and thiocyanate (1,4,11) anions, respectively. In addition, $\text{PPO}_8\text{CF}_3\text{SO}_3$ has been studied up to 180C and no SP was observed (11). This sequence is, in fact, in accord with the variation of the ion-ion interactions for these salts as reflected in the melting point. Specifically, the melting points for NaI , NaClO_4 , NaSCN , and NaCF_3SO_3 are 933 (12), 755 (12), 560 (13), and 520K (11). The idea, then, is that the higher the melting point, the larger the ion-ion interaction and consequently SP temperature should be lower. Finally, it is noted that this only holds for salts with a common cation. For example, KSCN exhibits a very low SP temperature 60C (3) and also has a very low melting point of 446K (13). The difference, of course, is due to the difficulty which the polymer has in solvating the large potassium ion. This serves to emphasize the other important aspect of this problem, namely, the ion-polymer interaction.

High Pressure Effects

Next, the effect of pressure on the electrical conductivity was determined and typical results are shown in Fig. 7. The slope of the curves at zero pressure were used to determine activation volumes directly via $\Delta V^* = -kT(d \ln \sigma / dP)$. The values of the activation volumes at 40, 60, 80, and 100C were found to be 67.9, 49.7, 41.5, and 25.4 cm^3/mol , respectively. These values are consistent with results for other ion conducting polymers and the large values are suggestive of cooperative molecular motions of polymer segments rather than single ion motions typical of framework type ion conductors where the activation volumes are about an order of magnitude smaller (14). In fact, the results are plotted in Fig. 8 along with other data for PPO

which exist including recent radio tracer results (15). (The glass transition for PPO_8NaSCN was taken to be 266K (11).) It is noted that these data are plotted vs. the "central" glass transition temperature which is different from previous plots by the authors which utilized T_0 (2,5) obtained from VTF fits to the data.

Previous high pressure NMR results for PPO_8NaI showed that the mobile ^{23}Na resonance in the narrowed regime (about 40C) is substantially broadened by an applied pressure of 2 kbar (3). The linewidth, $1/(T_2^*)$, of the mobile resonance in PPO_8NaI at 45C is plotted as a function of pressure in Fig. 9. (The lack of agreement between $1/(T_2^*)$ values at comparable temperatures and pressures from the data in Figs. 3 and 9 is attributable to magnetic field inhomogeneities associated with the high pressure NMR probe.) The simplest interpretation of the data is that the pressure induces an increase in T_g which, in turn, pushes the ^{23}Na linewidth back out of the motional narrowing regime. This result is consistent with previously observed shifts in T_g for uncomplexed PPO with pressure (5). Finally, the mobile/bound Na ratio for PPO_8NaI at 45C is plotted as a function of pressure in Fig. 10. The sample utilized in obtaining the data in Fig. 10 had previously been heated above 110C, and it is probable that not all of the precipitated salt had gone back into the complex by the time the high pressure measurements were performed. The decrease in M/B with increasing pressure can also be explained in terms of a T_g shift, as the data in Fig. 4 show that M/B exhibits an initial increase above T_g before precipitation effects occur.

SUMMARY

- in summary, then, the following results have been obtained.
- (a) PPO_nNaI samples are not homogeneous for $n > 8$. At low temperatures, at least, they consist of two well defined amorphous phases, one similar to uncomplexed PPO and another concentrated phase with n slightly smaller 8.
 - (b) The results imply that the concentrated phase is a conducting phase.
 - (c) For PPO NaI , the SP temperature increases as the concentration decreases.
 - (d) For various salts in PPO, it is found that the SP temperature increases as the ion-ion interaction to the extent that the ion-ion interaction is given by the melting point of the salt.
 - (e) There is no evidence for the presence of contact ion pairs or triplets, unless they exist on a timescale substantially shorter than that probed by the NMR measurements.
 - (f) The effects of pressure on the electrical conductivity and NMR have been determined and all results emphasize the role of the glass transition temperature concerning ion transport.

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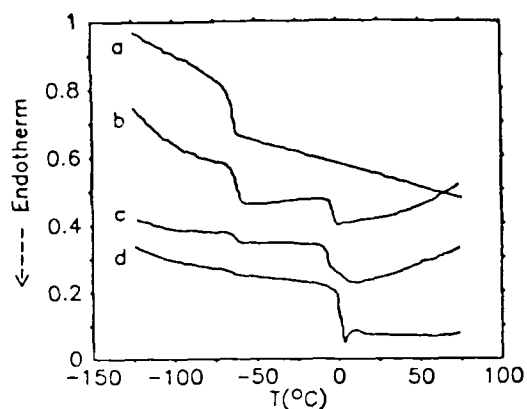


Figure 1a. DSC thermograms for: (a) uncomplexed PPO, (b) $\text{PPO}_{16}\text{NaI}$, (c) $\text{PPO}_{12}\text{NaI}$, and (d) PPO_8NaI . The data were taken at 10K/min.

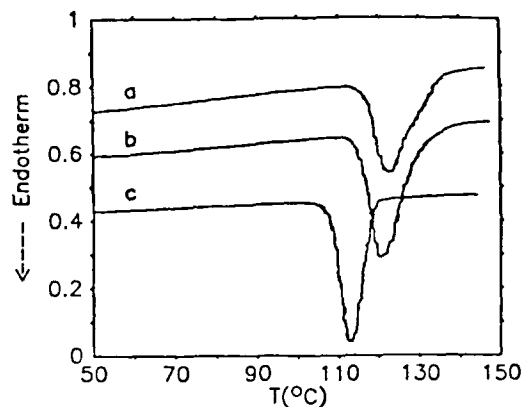


Figure 1b. DSC thermograms for: (a) $\text{PPO}_{16}\text{NaI}$, (b) $\text{PPO}_{12}\text{NaI}$, and (c) PPO_8NaI . The data were taken at 10K/min.

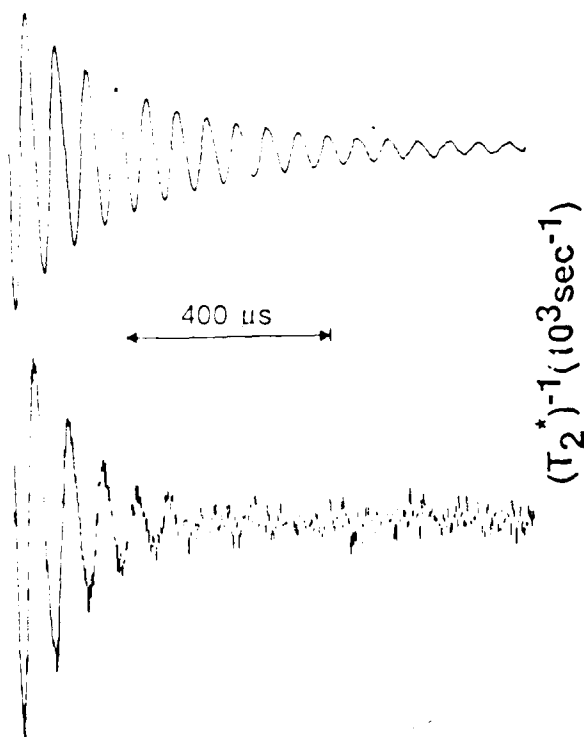


Figure 2. ^{23}Na free induction decays (FID's) for the mobile (top) and bound (bottom) Na in PPO_8NaI at 56°C. The vertical gain of the bottom FID is a factor of 3.3 higher than for the top.

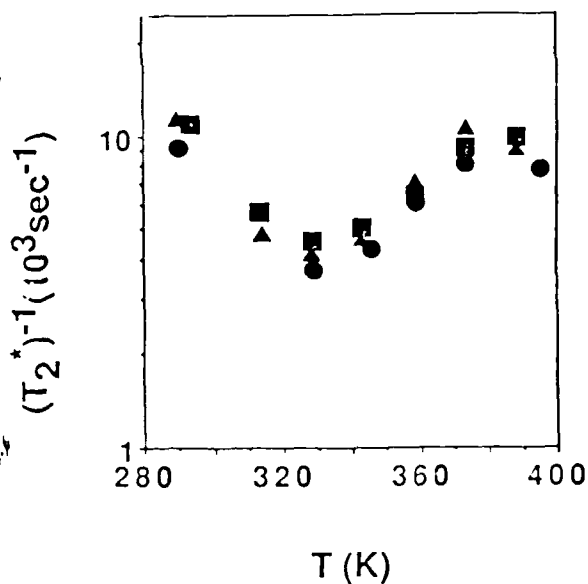


Figure 3. ^{23}Na reciprocal time constants $1/(T_2^*)$ of the mobile Na in PPO_nNaI ; $n=8$: circles, $n=12$: triangles, $n=16$: squares.

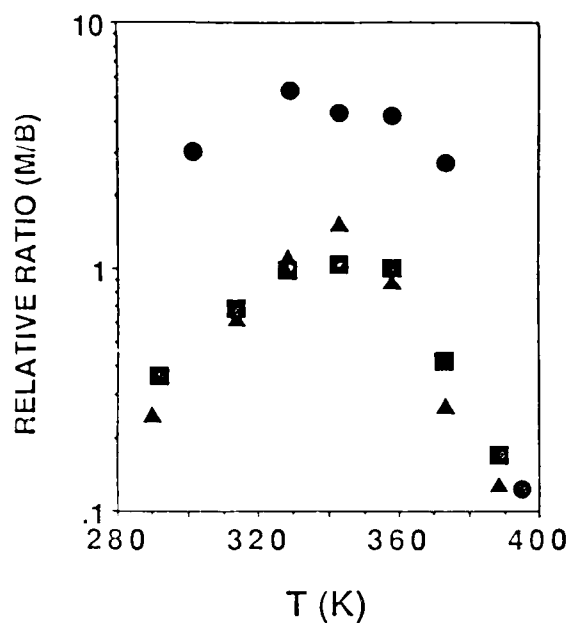


Figure 4. Mobile to bound Na ratios as a function of temperature, determined from FID intensities; $n=8$: circles, $n=12$: triangles, $n=16$: squares.

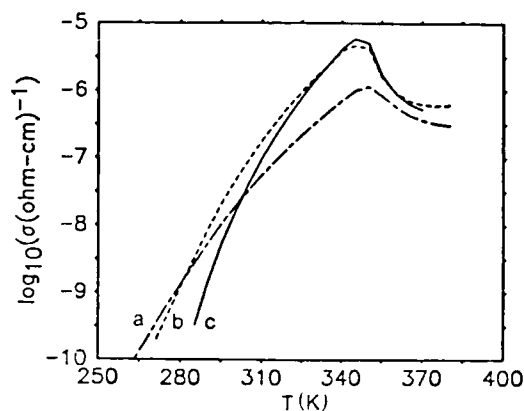


Figure 5. Electrical conductivity as a function of temperature. The curves are: (a) $\text{PPO}_{16}\text{NaI}$ (chain link), (b) $\text{PPO}_{12}\text{NaI}$ (dash), and (c) PPO_8NaI (solid). Straight line segments connect the datum points which are about 50 apart.

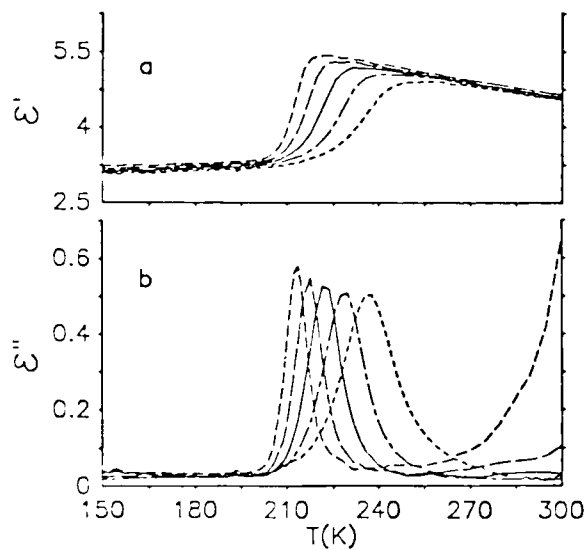


Figure 6. Real (a) and imaginary (b) parts of the dielectric constant at five frequencies; 10 Hz: short dash, 100 Hz: long dash, 1000 Hz: solid, 10,000 Hz: chain link, and 100,000 Hz: dotted lines.

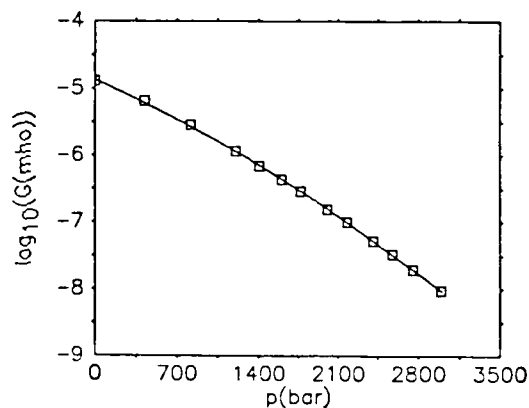


Figure 7. Electrical conductance (proportional to conductivity) as a function of pressure at 60°C for PPO_8NaI .

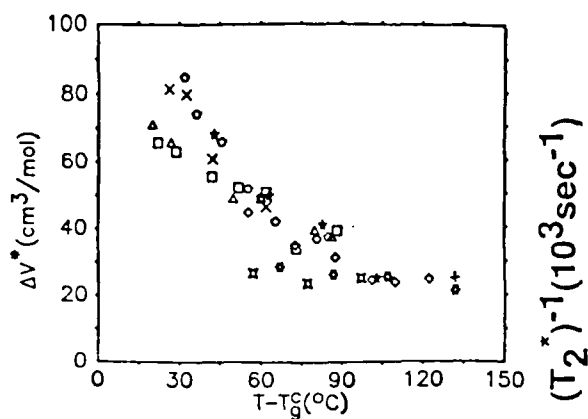


Figure 8. Activation volume vs. temperature relative to the glass transition; $\text{PPO}_8\text{LiCF}_3\text{SO}_3$: \diamond , $\text{PPO}_8\text{LiClO}_4$: \square , PPO_8LiI : Δ , uncomplexed PPO α relaxation time: \times , PPO_8LiSCN : \boxtimes , and $\text{PPO}_8\text{NaClO}_4$: \circ (2,6), PPO_8NaI : \star (present work), PPO_8NaSCN : \star (electrical conductivity, ref. 15), $+$ (^{14}C and ^{22}Na radio tracer, ref. 15).

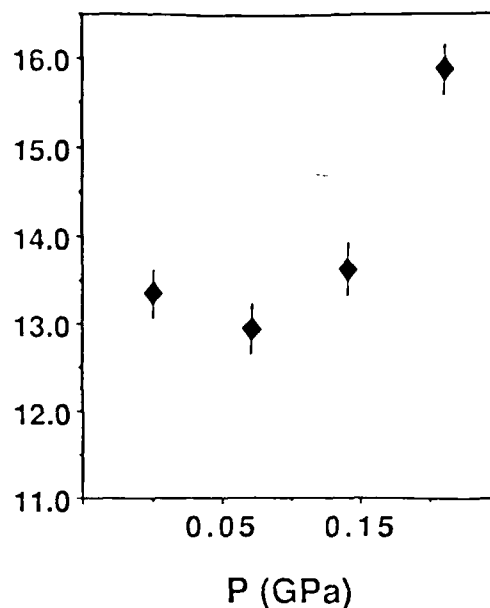


Figure 9. ^{23}Na reciprocal FID time constant $1/(T_2^*)$ of the mobile Na in PPO_8NaI at 45°C as a function of pressure.

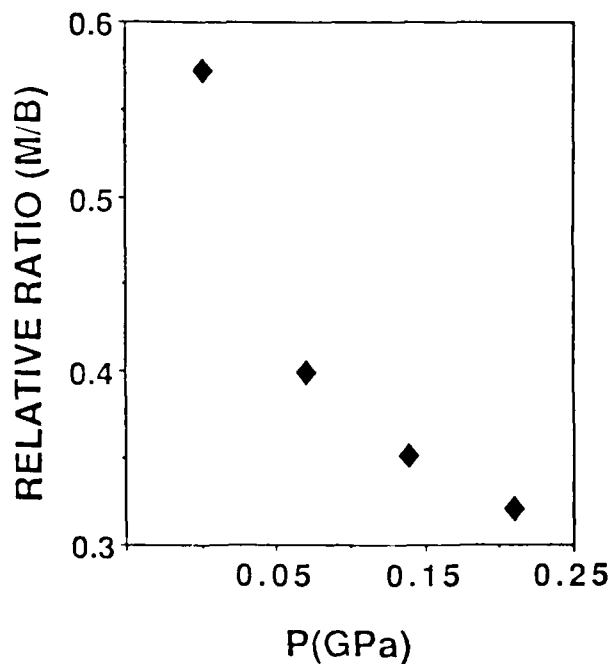


Figure 10. Mobile to bound Na ratio for PPO_8NaI at 45°C as a function of pressure.

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